Defense Technical Information Center Compilation Part Notice

ADP012393

TITLE: Atomic Fluorine Source for Chemical Lasers

DISTRIBUTION: Approved for public release, distribution unlimited

This paper is part of the following report:

TITLE: Gas and Chemical Lasers and Intense Beam Applications III Held in San Jose, CA, USA on 22-24 January 2002

To order the complete compilation report, use: ADA403173

The following component part numbers comprise the compilation report:
ADP012376 thru ADP012405

Atomic fluorine source for chemical lasers

S.J. Davis*, D.B. Oakes, M.E. Read, and A.H. Gelb
Physical Sciences Inc.

ABSTRACT

We present results from the early development of an F atom source appropriate for HF and AGIL chemical laser research. The system uses high power microwaves to produce a high enthalpy plasma that thermally dissociates molecular species such as SF_6 and F_2. Results of the characterization of the flow are presented.

Keywords: hydrogen fluoride lasers, combustors, halogen atom sources, microwave discharges

1. INTRODUCTION

Chemical lasers offer the obvious advantage that the population inversion is produced by selective chemical reactions, e.g., HF lasers[1] or energy transfer from chemically produced metastable species such as the Chemical Oxygen Iodine Laser (COIL)[2] and All Gas-Phase Iodine Laser (AGIL).[3-6] Often, however, one must supply energy sources to obtain the chemical species required for the selective reactions to occur. For example the reactants that produce the HF laser are F and H_2. Consequently, production of F atoms is a key element in HF chemical lasers. Several methods for F atom production have been used: DC electric discharge, photolyis, electrical arc heating, and chemical combustors. Large, high power HF lasers use chemical combustors to produce the enthalpy necessary to thermally dissociate either F_2 or NF_3. The combustors can be driven by the reaction of H_2 and F_2. For small, laboratory-scale HF lasers DC electric discharge is often used.

In this paper we discuss a new, alternate dissociation source for F atom production that may be suitable for HF and other chemical lasers that require F atoms such as HF and the new AGIL system. Our source is based on a device called the Microwave Induced Plasma Jet (MIDJet™). The MIDJet™ device can be considered to be a thermal dissociation source of atomic species. In effect, it is an efficient electrical combustor. The MIDJet™ is shown schematically in Fig. 1. It consists of a coaxial microwave line feeding a TM_{01} microwave cavity formed by an outer cylinder and an inner

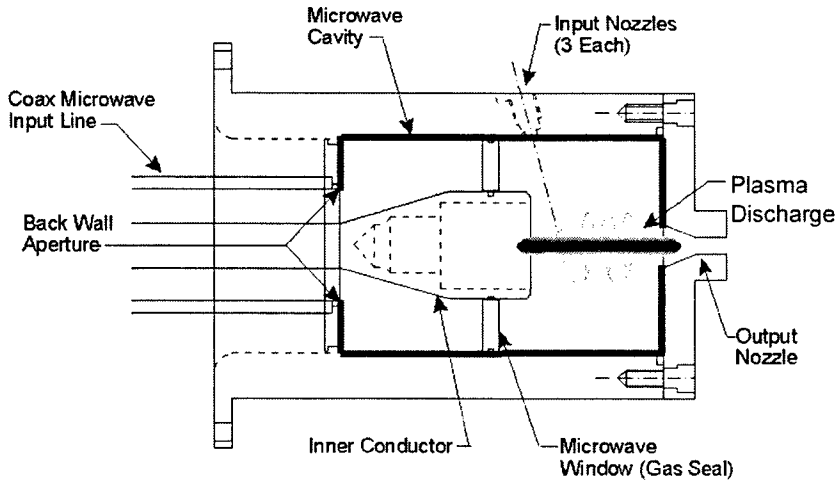

Fig. 1: Schematic of a MIDJet™ device.

*sdavis@psicorp.com; phone 1 978 689-0003; fax 1 978 689 3232; http://www.psicorp.com; Physical Sciences Inc., 20 New England Business Center, Andover, MA, USA 01810-1077

Gas and Chemical Lasers and Intense Beam Applications III, Steven J. Davis, Michael C. Heaven, Editors, Proceedings of SPIE Vol. 4631 (2002) © 2002 SPIE · 0277-786X/02/$15.00

conductor. Microwave power is injected via a coaxial line. The inner conductor is continued into the cavity, forming a coaxial structure that allows the diameter to be significantly smaller than would be possible with an open cavity. A discharge is produced on axis, where the RF electric field is locally maximum. The input gas is introduced via nozzles in the outer wall, with nozzles angled to produce an azimuthal (vortex) flow. This flow produces a radial acceleration that results in hot gas being pushed inward. The discharge produced by the high RF electric field on axis is thus confined to a small cylindrical volume. The gas exits through a nozzle. The output flow can be subsonic, sonic, or supersonic, depending on the pressure difference between the torch and the ambient environment, and the design of the nozzle. The MIDJet™ can be operated with internal pressures from slightly sub-atmosphere to more than 10 atm, producing supersonic and subsonic flows even into the atmosphere.

2. EXPERIMENT

We assembled the apparatus shown in Fig. 2 to test the MIDJet™ as an F-atom source appropriate for chemical laser research. The MIDJet™ head was connected to a chamber with three pumping stages. The chamber was a six port cross with each port 20 cm in diameter. As indicated in Fig. 2, for some tests we used a mass spectrometer to analyze the effluents from the MIDJet™ head to measure the dissociation fraction of the molecular fluorine. This spectrometer can make measurements up to 300 amu with 1 amu resolution. Two stages of differential pumping are used to handle the high mass flow rates of the MIDJet™. Greater than 99% of the flow is pumped from the first stage by a 400 cfm Roots Blower. A 0.5 mm diameter graphite skimmer is used to separate the first pumping stage from the second. This skimmer is well downstream of the Mach disk which forms about 2 cm from the MIDJet™ exit nozzle. Beyond the Mach disk the flow is subsonic. A second skimmer separates the second differential chamber from the first and provides the entrance path for gas to be analyzed by the mass spectrometer.

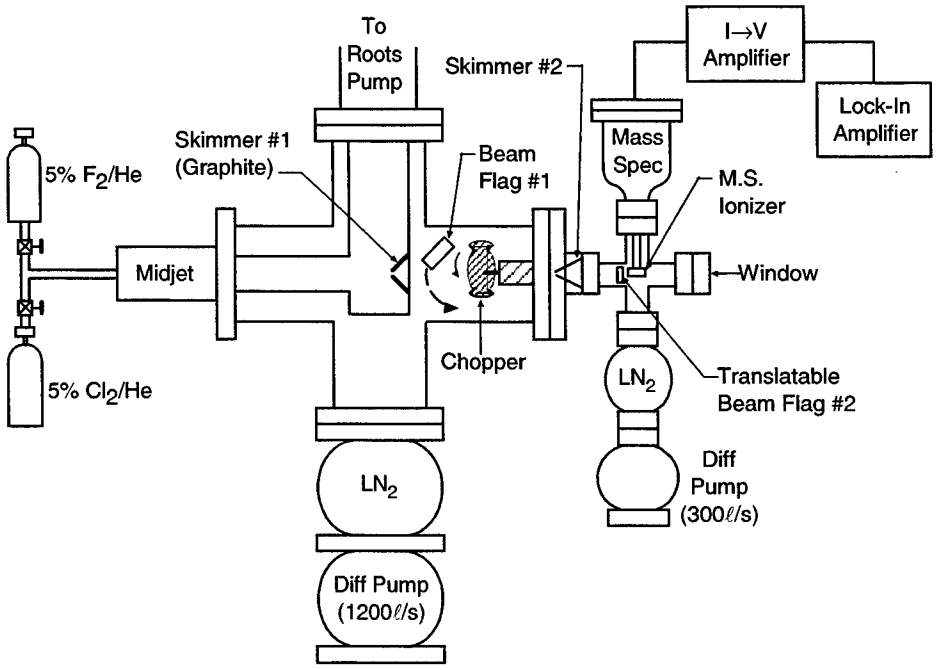

Fig. 2: Schematic of the apparatus used for the beam composition measurements.

The beam composition is measured by monitoring the mass 19 (F atom) and 38 (F_2) signals. To convert the raw signals into values that are proportional to the concentration of F and F_2 in the flow we must take into account the following: 1) F signal resulting from F_2 cracking in the mass spectrometer ionizer, 2) the relative ionization cross section by electron beam impact for F and F_2, and 3) the transmission function (sensitivity versus mass) of the mass spectrometer. For the fluorine beam the atomic content of the fluorine component of the beam (%F, neglecting the helium diluent) is given by Eq. (1).

$$\% \text{ F atoms} = \frac{N_F}{(N_F + N_F 2)} \times 100\% \quad (1)$$

where:

$$N_F = \left(S_F - S_F^{F2}\right) T_F^{-1} C_F^{-1}$$
$$N_{F2} = S_{F2} T_{F2}^{-1} C_{F2}^{-1} \quad (2)$$

and:

N_F = Value proportional to # of F atoms in beam
N_{F2} = Value proportional to # of F_2 molecules in beam
S_F = The m/e = 19 signal (Volts)
S_{F2} = The m/e = 38 signal (Volts)
S_F^{F2} = The contribution to the m/e = 19 signal due to the cracking of F_2
T_F = The transmission function for m/e = 18
T_{F2} = The transmission function for m/e = 38
C_F = The relative ionization cross section for atomic fluorine
C_{F2} = The relative ionization cross section for F_2.

We obtained references reporting measurements of the absolute cross section for ionization of F and F_2 by electron impact at 70 eV (our electron energy).[7,8] We also measured the relative transmission function for the mass spectrometer from mass (m/e) 20 to 40. The relative Ar^+ (m/e = 40) and Ar^{++} (m/e = 20) signals in our mass spectrometer formed by electron impact of argon were compared to the published cross section for the formation of Ar^+ and Ar^{++} by electron impact of argon. This comparison led to values of the relative transmission function between mass 19 and 38. Finally the contribution of F_2 cracking to the F signal was measured by turning the MIDJet™ plasma off and measuring the relative m/e = 38 to 19 signals for the cold flow. A summary of all the parameters necessary for the calculation of the atomic composition of the fluorine beam is presented in Table 1.

Table 1: Summary of the Parameters Used to Determine the Fluorine Beam Composition

Beam	Transmission Function (Relative)		Ionization Cross Section (x 10^{-16} cm^2)		Fraction of Parent Signal that Contributes to Atom Signal
	Atom	Parent	Atom	Parent	
Fluorine	1.00	0.763	0.87	1.10	0.210

3. RESULTS

MIDJet™ beams were made from the 5%F_2/Helium mixture with discharge powers ranging from 1860 to 3610 W. The results of the beam composition analysis are plotted as % F_2 dissociation versus Power/Flow ratio in Fig. 3.

Fig. 3 shows that the maximum F atom content of the fluorine flow obtained in these experiments was 81%. Equilibrium calculations predict that the fluorine in the 5% F_2/He beam will be essentially fully dissociated for Power/Flow ratios greater than 40 W/slm. We are therefore observing less dissociation than expected based upon the equilibrium prediction. However, as expected, at a fixed discharge power the data does show a correlation between F atom content and increasing Power/Flow ratio.

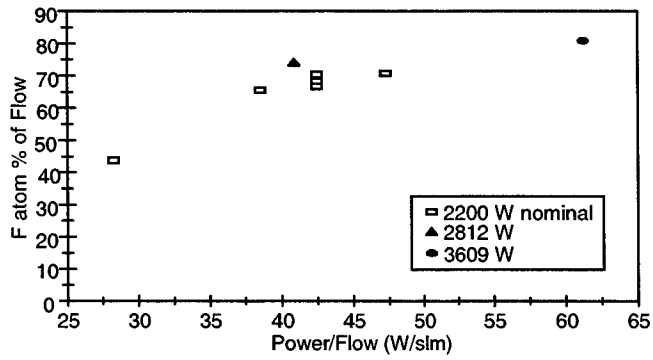

Fig. 3: F atom % of the flow versus Power/Flow (W/slm) ratio for the beams formed from the 5% F_2/helium mixture.

We estimated that the effects of gas phase recombination in the chamber are negligible. However, recirculation flows which result in background gas reaching and passing through the first skimmer would have a high recombination fraction due to substantial interaction with the chamber walls. A summary of a few composition measurements are shown in Table 2.

Table 2: Summary of the Conditions and Results for Three of the Composition Experiments Included in Fig. 3

Coupled Discharge Power (W)	Power/Flow (W/slm)	Formation Chamber Pressure (Torr)	Atomic F % of Flow	F Atom Fluence (/s)	F Atom Fluence (mmoles/s)
1880	42.5	2.80	68.2	1.02×10^{21}	1.8
2810	40.9	4.05	74.2	1.82×10^{21}	3.0
3610	61.2	3.67	80.7	1.79×10^{21}	3.0

Since the MIDJet™ device can operate at pressures of an atmosphere or greater, it is appropriate to consider it as a driver for a subsonic or supersonic chemical laser nozzle. One could consider the downstream side of the device as a plenum. The gases would then flow through a mixing nozzle as indicated in Fig. 4.

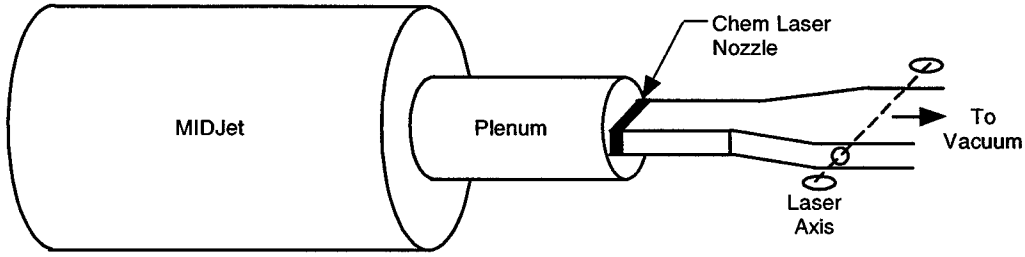

Fig. 4: Concept for a MIDJet™ driven chemical laser.

3.1 Scaled MidJet™

We have designed and are building a scaled device that will deliver 25 kW of microwave power to the flow. Equilibrium calculations indicate that such as system will be able to produce nearly 50 mmoles of F atoms. For example, in Figs. 5 and 6 we show results from an equilibrium calculation for a flow of 16 mmoles/s of F_2 and NF_3 in a flow of 700 mmoles of He. The equilibrium calculations use the heat capacities of the gases and predict dissociation fractions as a function of temperature. Fig. 5 indicates that 25 kW of absorbed power will dissociate greater than 95% of the F_2 in the flow. For NF_3 (see Fig. 6), greater than 90% of the NF_3 will be dissociated into F atoms.

The expected dissociation products from NF_3 are indicated in Fig. 7.

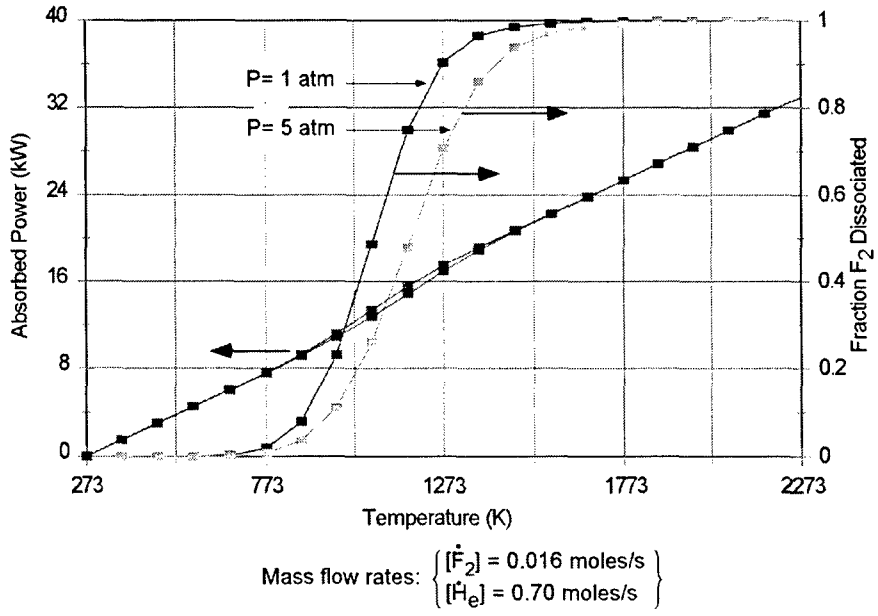

Fig. 5: Equilibrium code predictions of dissociation fraction of F_2 for a mix of F_2/He.

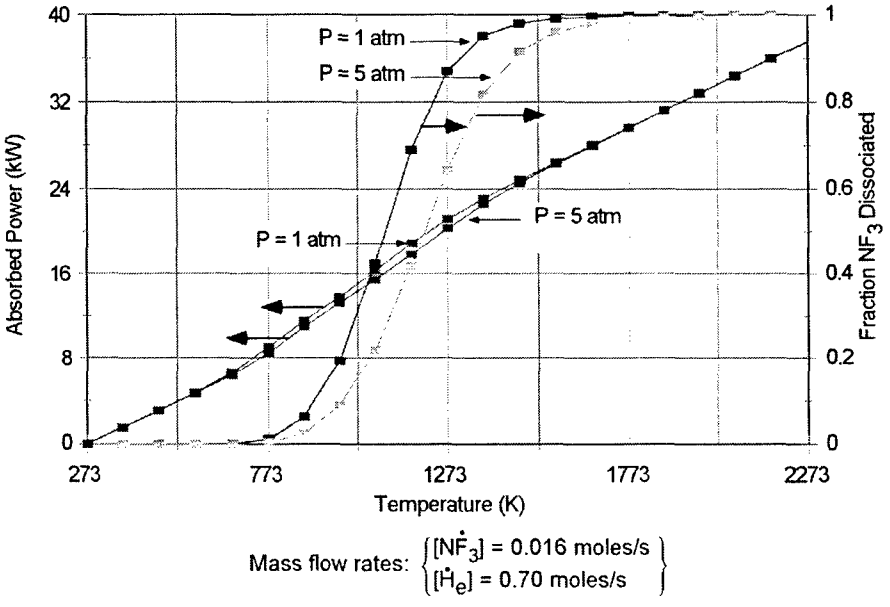

Fig. 6: Equilibrium code prediction of dissociation fraction of NF_3 for a mix of NF_3/He.

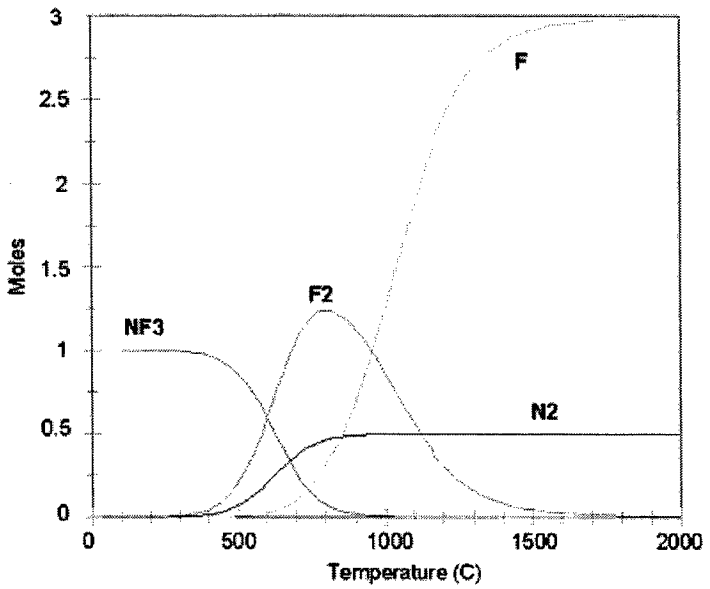

Fig. 7: Equilibrium calculation for NF_3 at 1 atm.

4. SUMMARY

We have described a novel, electrically based source for halogen atoms that may be a convenient source of F atoms for HF chemical laser research. We have characterized a device that delivers up to 3.5 kW to the gas flow and have demonstrated F atom fluences of 3 mmoles/s for a 5% mix of F_2 in He. A scaled device is under construction.

ACKNOWLEDGMENT OF SUPPORT AND DISCLAIMER

This material is based upon work supported by the United States Air Force Research Laboratory, Directed Energy Directorate, Contract No. F29601-00-C-0053. We gratefully acknowledge this support.

REFERENCES

1. R.W.F. Gross and J.F. Bott, *Handbook of Chemical Lasers*, John Wiley & Sons, New York, 1976.
2. W.E. McDermott, N.R. Pchelkin, D.J. Benard, and R.R. Bousek, "An Electronic Transition Chemical Laser," *Appl. Phys. Lett.*, **32**, 469, 1978.
3. J.M. Herbelin, T.L. Henshaw, B.D. Rafferty, B.T. Anderson, R.F. Tate, T J. Madden, G.C. Manke II, and G.D. Hager, "The Measurement of Gain on the 1.315 μm Transition in Atomic Iodine in a Subsonic Flow of Chemically Generated $NCl(a^1\Delta)$," *Chem. Phys. Lett.,*. **299**, 583, 1999.
4. T.T. Yang, V.T. Gylys, R.D. Bower, and L.F. Rubin, "Population Inversion Between $I(^2P_{1/2})$ and $I(^2P_{3/2})$ of Atomic Iodine Generated by Excitation Transfer from $NCl(a^1\Delta)$ to $I(^2P_{3/2})$," *SPIE*, **1871,** 1993.
5. A.J. Ray and R.D. Coombe, "Energy Transfer from $NCl(a^1\Delta)$ to Iodine Atoms,", *J. Phys. Chem.*, **97**, 3475, 1993.
6. A.J. Ray and R.D. Coombe, "An I^* Laser Pumped with $NCl(a^1\Delta)$," *J. Phys. Chem.*, **99**, 7849, 1995.
7. T.R. Hayes, R.C. Wetzel, and R.S. Freund, "Absolute Electron-Impact-Ionization Measurements of the Halogen Atoms," *Phys. Rev. A*, **35**(2), 578, 1987.
8. F.A. Stevie and M.J. Vasile, "Electron Impact Ionization Cross Sections of F_2 and Cl_2," *J. Chem. Phys.*, **74**(9), 5106, 1981.